

# A first-principles model of spectrally resolved 5.3 $\mu\text{m}$ nitric oxide emission from aurorally dosed nighttime high-altitude terrestrial thermosphere

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[1] The spectrally resolved nighttime 5.3  $\mu\text{m}$  emission from NO observed by the Cryogenic Infrared Radiance Instrumentation for Shuttle (CIRRIS-1A) experiment aboard space shuttle Discovery at 195 km tangent altitude during a strong auroral event is modeled using a first-principles kinetics model. An appropriate SHARC (Strategic High Altitude Radiance Code) Atmospheric Generator (SAG) is dosed with an IBC class III aurora. The spectrally resolved fundamental vibration-rotation band emissions from NO around 5.3  $\mu\text{m}$  resulting from impacts of ambient NO with O as well as reactions of N atoms with O<sub>2</sub> are calculated under steady state conditions. The calculated results, using a local translational temperature derived from the observed spectrum, are in excellent agreement with the CIRRIS-1A observations, validating our model. The importance of the accurate nascent vibrational and rotational distribution of chemically produced NO as well as the collisionally induced rotation-to-vibration relaxation of rotationally hot NO is pointed out. **Citation:** Duff, J. W., H. Dothe, and R. D. Sharma (2005), A first-principles model of spectrally resolved 5.3  $\mu\text{m}$  nitric oxide emission from aurorally dosed nighttime high-altitude terrestrial thermosphere, *Geophys. Res. Lett.*, 32, L17108, doi:10.1029/2005GL023124.

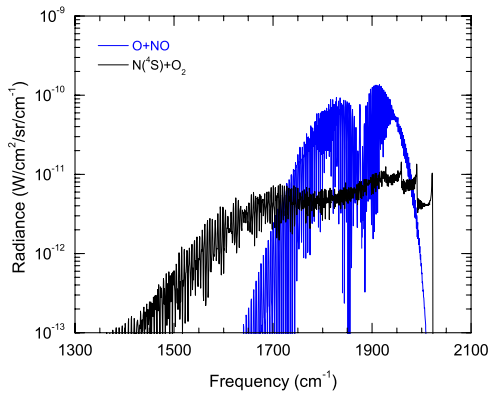
## 1. Introduction

[2] The 5.3  $\mu\text{m}$  fundamental vibration-rotation band ( $\Delta\nu = -1$ ;  $\Delta j = 0, \pm 1$ ) emission from NO is the strongest cooling agent in the 110–300 km region of the terrestrial thermosphere [Kockarts, 1980; Zachor *et al.*, 1985; Sharma *et al.*, 1998; Sharma and Roble, 2001]. The 5.3  $\mu\text{m}$  emission from the fundamental band as well as that from the overtone band ( $\Delta\nu = -2$ ;  $\Delta j = 0, \pm 1$ ) of NO around 2.7  $\mu\text{m}$  are both greatly enhanced during an auroral event [Sharma *et al.*, 2000, 2001]. This is dramatically illustrated by the global images of 5.3  $\mu\text{m}$  emission from NO taken by SABER (Sounding of the Atmosphere using Broadband Emission Radiometry) instrument aboard NASA's TIMED (Thermosphere-Ionosphere-Mesosphere Energetics and Dynamics) satellite on the 15 and 19 April 2002, before and after solar coronal mass ejection events, respectively [Russell *et al.*, 2002]. Mlynczak *et al.* [2003] point out that “the 5.3  $\mu\text{m}$  limb

radiance increases by a factor 5 to 7 during the storm time, and the altitude range over which the radiance is recorded increases by nearly 45 km,” from 280 to 325 km tangent altitude. Since these emissions from NO are an important factor in determining the temperature and density structure of the atmosphere [Sharma and Roble, 2001], it is important that they be quantitatively modeled. In this letter we reexamine the model of the spectrally resolved emission from NO around 5.3  $\mu\text{m}$  observed at 2293 LT on 29 April 1991 by the space shuttle experiment CIRRIS-1A for a tangent altitude of 195 km during a strong (IBC III) auroral event. In the previous effort to model this aurora [Dothe *et al.*, 2002] we were limited by the lack of knowledge of the temperature dependence of the rate coefficient of the  $\text{N}(^2\text{D}) + \text{O}_2 \rightarrow \text{NO} + \text{O}$  reaction as well as the vibration-rotation distribution of the product NO. Duff *et al.* [2003] have since shown that this rate coefficient is given by the expression  $k(T) = 6.2 \times 10^{-12} (T/300) \text{ cm}^3/(\text{s-molecule})$  instead of the nearly temperature independent value  $2.8 \times 10^{-12} \exp(185/T) \text{ cm}^3/(\text{s-molecule})$  given in the literature [Herron, 1999]. Sharma *et al.* [2002] have in addition reported the vibrational-rotation distribution of the nascent NO produced by this reaction at room temperature.

[3] Figure 1 plots the calculated nighttime fundamental vibration-rotation NO band radiance ( $\text{Watts}/(\text{cm}^2\text{-sr-cm}^{-1})$ ) as a function of frequency from a quiescent atmosphere at 195 km tangent altitude. The width of the P and R branches of the spectrum resulting from the inelastic impacts of ambient NO( $v = 0$ ) with O (blue curve) indicates low rotational temperature. The black curve is the chemiluminescent emission resulting from the nascent NO formed by the reaction  $\text{N}(^4\text{S}) + \text{O}_2 \rightarrow \text{NO} + \text{O}$ . This reaction is exothermic by 1.385 eV but has activation energy of about 0.3 eV [Duff *et al.*, 1994]. However, at the local translational temperature of 1065 K (0.092 eV) a significant fraction of atom-molecule pairs in the tail of the Maxwell-Boltzmann distribution have relative translational energy to overcome the barrier. Three R-branch bandheads, for  $1 \rightarrow 0$ ,  $2 \rightarrow 1$ , and  $3 \rightarrow 2$  vibrational transitions, are clearly identified and the fourth one for the  $4 \rightarrow 3$  vibrational transition near  $1920 \text{ cm}^{-1}$  is apparent. Recalling that the rotational levels contributing to the bandheads have rotational energy in excess of 1 eV and noting the width of the emission envelope, one concludes that average rotational energy of the nascent NO produced by this reaction is at least several thousand degrees K. The emission due to

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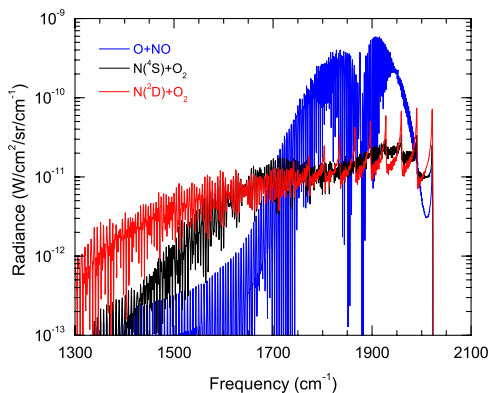
**Figure 1.** Contribution of the  $\text{O} + \text{NO}(v=0) \rightarrow \text{NO}(v'=1) + \text{O}$  vibrational excitation (blue curve) and the  $\text{N}(^4\text{S}) + \text{O}_2 \rightarrow \text{NO}(v) + \text{O}$  chemiluminescent reaction (black curve) to the ambient model spectrum.

$\text{N}(^2\text{D}) + \text{O}_2 \rightarrow \text{NO} + \text{O}$  is not seen because of the absence of  $\text{N}(^2\text{D})$  atoms in the quiescent nighttime atmosphere, a conclusion supported by *Sharma et al.* [1996].

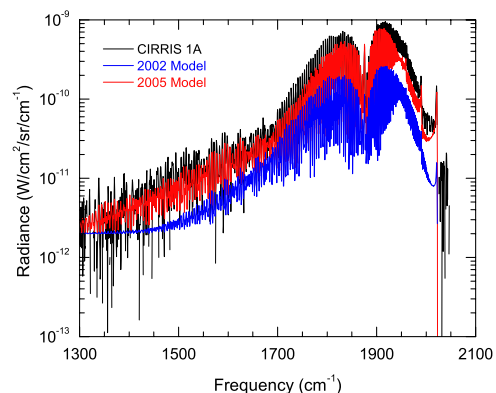
[4] Figure 2 plots the calculated nighttime fundamental vibration-rotation NO band radiance ( $\text{Watts}/(\text{cm}^2\text{-sr-cm}^{-1})$ ) as a function of frequency from an IBC III aurorally dosed atmosphere at 195 km tangent altitude. Again, the blue curve is the emission resulting from the inelastic impacts, including pure rotational relaxation and vibrational rotational excitation, of ambient  $\text{NO}(v=0, j)$  with O. The vibrational-rotational and pure rotational relaxation rate constants for  $\text{O} + \text{NO}(v, j)$  are based on quasi-classical trajectory calculations [*Duff et al.*, 1998] using information developed previously for  $\text{O} + \text{NO}(v)$  vibrational relaxation [*Duff and Sharma*, 1997]. The radiance level as well as the width of the P and R branches of the spectrum is larger than for the quiescent case (Figure 1) indicating a higher rotational temperature. This is because the reaction of  $\text{N}(^4\text{S})$  and  $\text{N}(^2\text{D})$  atoms with  $\text{O}_2$  produce highly rotationally and vibrationally excited NO [*Duff et al.*, 1994; *Sharma et al.*, 2002] which, after losing the vibrational energy, is still highly rotationally excited. These highly rotationally

excited NO molecules have a larger cross section for vibrational excitation upon impacts with O and yield vibrationally excited NO molecules with higher rotational temperature. The black curve is the chemiluminescent emission resulting from the nascent NO formed by the reaction  $\text{N}(^4\text{S}) + \text{O}_2 \rightarrow \text{NO} + \text{O}$ . The radiance level is enhanced over the quiescent case because of the production of  $\text{N}(^4\text{S})$  atoms by auroral dosing. The width of the emission spectrum is about the same as for the quiescent case indicating that energy going into the rotational degree of freedom is about the same. The red curve is the chemiluminescent emission resulting from the nascent NO formed by the reaction  $\text{N}(^2\text{D}) + \text{O}_2 \rightarrow \text{NO} + \text{O}$ . This reaction is exothermic by 3.76 eV, has no activation energy and produces NO in high rotational and vibrational levels. The rate coefficient of this reaction as function of rotational and vibrational quantum numbers of the nascent NO has been presented previously [*Duff et al.*, 2002; *Sharma et al.*, 2002]. The highly vibrationally and rotationally excited nascent NO molecules from the  $\text{N}(^2\text{D}) + \text{O}_2$  reaction lead, as shown by the red curve in Figure 2, to a few R-branch bandheads and emission over a large spectral range—a result of the calculated rotational temperature of the radiatively relaxed  $\text{NO}(v)$  produced by the  $\text{N}(^2\text{D}) + \text{O}_2$  reaction varying with the vibrational level  $v$ , 8400 K for  $v=1$  to 4400 K for  $v=12$ .

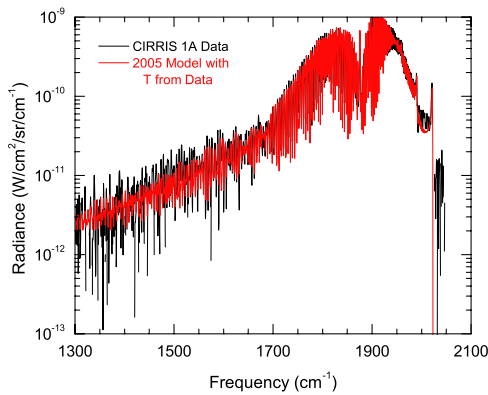
[5] Figure 3 compares the CIRIS-1A data and the sum of the curves of Figure 2 as well as the “standard model” [*Dothe et al.*, 2002]. Larger amounts of NO produced over the “standard model,” because of the approximately three times faster  $\text{N}(^2\text{D}) + \text{O}_2$  rate coefficient of *Duff et al.* [2003] and thus a faster rate of excitation of rotationally hot  $\text{NO}(v=0)$  by impacts with atomic oxygen, lead to a significant improvement in the agreement of the model with the CIRIS-1A observations in the 1800–2000  $\text{cm}^{-1}$  region of the spectrum. Furthermore, the detailed treatment (including vibrational-rotational excitation/de-excitation and radiative relaxation) of rotationally hot  $\text{NO}(v)$  from the  $\text{N}(^2\text{D}) + \text{O}_2$  reaction (average rotational temperature of  $\sim 5500$  K) also leads to excellent agreement between the data and present model in the 1400–1700  $\text{cm}^{-1}$  spectral region. The faster rate of excitation of rotationally hot



**Figure 2.** Contribution of the  $\text{O} + \text{NO}(v=0) \rightarrow \text{NO}(v'=1) + \text{O}$  vibrational excitation (blue curve), the  $\text{N}(^4\text{S}) + \text{O}_2 \rightarrow \text{NO}(v', T_{\text{rot}}) + \text{O}$  (black curve) and  $\text{N}(^2\text{D}) + \text{O}_2 \rightarrow \text{NO}(v', j') + \text{O}$  (red curve) chemiluminescent reactions to the current model spectrum assuming an IBC III aurora.



**Figure 3.** Calculated model spectrum (red curve) using the MSIS temperature of 1065 K and the observed auroral spectrum (black curve). Also shown is the “standard model” calculated spectrum (blue curve) [*Dothe et al.*, 2002].



**Figure 4.** Calculated model spectrum (red curve) assuming a translational temperature of 1160 K derived from the CIRRIS-1A measurement and the observed auroral spectrum (black curve).

$\text{NO}(v=0)$  by impacts with atomic oxygen has implications for the remote sensing of NO by inversion of its 5.3  $\mu\text{m}$  emission – an issue which is currently being investigated.

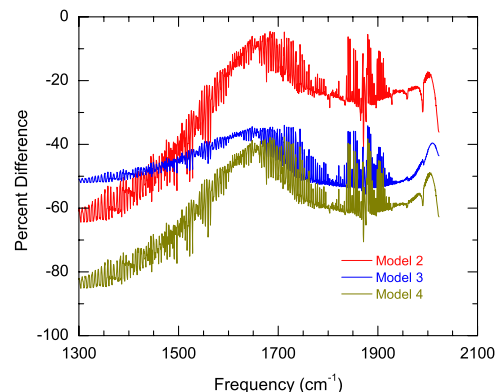
[6] The comparison shown in Figure 3 indicates that the model temperature (or spectral width) of the  $\text{NO}(v=1)$  thermal emission in the 1800–2000  $\text{cm}^{-1}$  spectral region is lower (or narrower) than that suggested by the observation. Our previous analysis [Dothe *et al.*, 2002] of the CIRRIS 1A spectrum indicated a rotational temperature of  $925 \pm 25$  K for the thermal  $\text{NO}(v=1 \rightarrow v=0)$  emission. Using the correlation between the temperature of the nascent  $\text{NO}(v=1)$  rotational distribution and translational temperature developed previously [Sharma and Duff, 1997] implies a translational temperature approximately 25–30% higher than the rotational temperature. Extending our previous results shows that a  $\text{NO}(v'=1, j')$  rotational distribution resulting from  $\text{NO}(v=0, T)$  collisions with O in a thermal bath of  $T = 1160$  K can be well represented by a Maxwell-Boltzmann distribution with a temperature of  $925 \pm 25$  K. The derived translational temperature of  $1160 \pm 30$  K at an altitude of 195 km is approximately 100 K greater than the MSIS temperature used in our previous and present models. Previous observations have also shown measured exospheric temperatures to be systematically higher than those predicted by MSIS during major geomagnetic storms [Dymond *et al.*, 2004].

[7] It should be noted that the data obtained by CIRRIS-1A is of high quality. Together with the earlier work [Sharma *et al.*, 1996, 1998] we estimate the signal-to-noise ratio in the observed spectra of 2.5–3 around 1400  $\text{cm}^{-1}$ .

[8] Figure 4 compares the CIRRIS 1A observation with the present model using a translational temperature of 1160 K. The remaining discrepancy between the present model and CIRRIS 1A observation (Figure 3) in the 1800–2000  $\text{cm}^{-1}$  spectral region is now removed, resulting in excellent agreement between the model and observation throughout the spectral range. This agreement requires a consistency between the atmospheric temperature,  $\text{O} + \text{NO}(v=0, j)$  collisional excitation/de-excitation rate coefficients, and the rate coefficients for the NO vibrational-rotational distributions resulting from the reactions of  $\text{N}(^4\text{S})$  and  $\text{N}(^2\text{D})$  atoms with  $\text{O}_2$ .

[9] Figure 5 plots percent difference ( $=[(\text{model } x - \text{model } 1)/\text{model } 1] \times 100$ ,  $x = 2, 3$ , and 4) between the current (model 1) with three other models. Model 2 and model 4 are the calculations using the MSISE-90 model atmosphere [Hedin, 1991] and NRLMSIS-00 [Picone *et al.*, 2002], respectively, together with the temperature independent  $\text{N}(^2\text{D}) + \text{O}_2$  rate coefficient used recently by Siskind *et al.* [2004] to analyze rocket data. Model 2 differs from the “standard model” of Dothe *et al.* [2002] in that the latter uses the  $\text{N}(^2\text{D}) + \text{O}_2$  rate coefficient recommended by Herron [1999]. Model 3 is the calculation using NRLMSIS together with the temperature dependent rate coefficient of Duff *et al.* [2003]. For Models 2 and 4, one would expect a decrease in radiance of a factor of about 4 around the band origin due to the decrease in the  $\text{N}(^2\text{D}) + \text{O}_2$  chemiluminescence. However, the less than a factor of 4 decrease is due to the increased importance of the  $\text{N}(^2\text{D}) + \text{O}$  quenching process compared to  $\text{N}(^2\text{D}) + \text{O}_2$  reaction (which is now slower by a factor of about 4). This increases the amount of  $\text{N}(^4\text{S})$  and thus the importance of the  $\text{N}(^4\text{S}) + \text{O}_2$  reaction. This is most important in the 1600–2000  $\text{cm}^{-1}$  frequency range, where  $\text{N}(^4\text{S}) + \text{O}_2$  reaction was previously of equal importance to (and is now of greater importance than) the  $\text{N}(^2\text{D}) + \text{O}_2$  reaction. In the 1300–1600  $\text{cm}^{-1}$  frequency range, the decrease in radiance is less due to the greater importance of the  $\text{N}(^4\text{S}) + \text{O}_2$  reaction. It should be noted that the radiance calculated by models 2 and 4 differs uniformly by a factor roughly equal to the ratio of the  $\text{O}_2$  density used in the two models. Model 3 differs uniformly from model 1 by a factor of about 2, again by a factor almost equal to the density ratio of  $\text{O}_2$  used in the two models.

[10] The temperatures at an altitude of 195 km from both MSISE-90 ( $= 1065$  K) and NRLMSISE-00 ( $= 1043$  K) are both lower than the 1160 K temperature deduced from the CIRRIS-1A interferometer data. This affects mostly the NO fundamental emission due to the  $\text{O} + \text{NO}(v=0)$  excitation mechanism, with a smaller effect around 1300–1700  $\text{cm}^{-1}$ . Although the NRLMSISE-00  $\text{O}_2$  number density is a factor of 2.4 lower than MSISE-90, it is to be recalled that the strength of the aurora obtained from our analysis of the  $\text{NO}(\Delta v=2)$  2.7  $\mu\text{m}$  radiometer data was based on the MSISE-90 atmosphere. If we used the lower  $\text{O}_2$  density from NRLMSISE-00, then we would have estimated a



**Figure 5.** Percent difference ( $=[(\text{model } x - \text{model } 1)/\text{model } 1] \times 100$ ,  $x = 2, 3$ , and 4) in the calculated radiances using different models as a function of frequency.



stronger aurora to compensate for the smaller production of NO.

[11] The earlier work of Dothe *et al.* [2002] showed that the 2002 model which assumes rotational LTE for NO produced by the  $\text{N}(\text{}^2\text{D}) + \text{O}_2$  reaction, even after being multiplied by a factor of three underestimates the observed radiance around  $1400\text{ cm}^{-1}$  by a factor of about 20. The current model using calculated high rotational temperatures gives excellent agreement with the radiance observed, not only in magnitude, but also in spectral shape. Since the vibrational transition probabilities in the first order are independent of the rotational quantum number, a change in the rotational temperature will just lead to increased in radiance in one part of the spectrum at the expense of another part. A quantitative assessment of the sensitivity of the observed spectrum to the rotational temperatures of different vibrational levels will be given in a later publication.

[12] In conclusion, the present model reproduces the CIRRIIS-1A observation of strong nighttime aurora with a high degree of fidelity. It underscores the need to know both the rotational and vibrational distributions of the nascent NO produced by the chemiluminescent reactions to correctly model fundamental vibration-rotation band emission from NO during an auroral event.

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